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**Key Points:**
- The effect of dam removal on riparian zone nitrogen (N) cycle processes and groundwater N concentrations is unknown
- We studied changes in riparian N cycle processes associated with the removal of a 1.5 m milldam for 2 years
- While soil denitrification and $\delta^{15}$N decreased following dam removal, an increase in groundwater nitrate-N concentrations was not observed

**Supporting Information:**

Supporting Information may be found in the online version of this article.

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# Draining the Landscape: How Do Nitrogen Concentrations in Riparian Groundwater and Stream Water Change Following Milldam Removal?

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**Abstract** Dam removals are on the increase across the US with Pennsylvania currently leading the nation. While most dam removals are driven by aquatic habitat and public safety considerations, we know little about how dam removals impact water quality and riparian zone processes. Dam removals decrease the stream base level, which results in dewatering of the riparian zone. We hypothesized that this dewatering of the riparian zone would increase nitrification and decrease denitrification, and thus result in nitrogen (N) leakage from riparian zones. This hypothesis was tested for a 1.5 m high milldam removal. Stream, soil water, and groundwater N concentrations were monitored over 2 years. Soil N concentrations and process rates and $\delta^{15}$N values were also determined. Denitrification rates and soil $\delta^{15}$N values in riparian sediments decreased supporting our hypothesis but no significant changes in nitrification were observed. While surficial soil water nitrate-N concentrations were high (median 4.5 mg N $L^{-1}$), riparian groundwater nitrate-N values were low (median 0.09 mg N $L^{-1}$), indicating that nitrate-N leakage was minimal. We attribute the low groundwater nitrate-N to denitrification losses at the lower, more dynamic, groundwater interface and/or dissimilatory nitrate reduction to ammonium (DNRA). Stream water nitrate-N concentrations were high (median 7.6 mg N $L^{-1}$) and contrary to our dam-removal hypothesis displayed a watershed-wide decline that was attributed to regional hydrologic changes. This study provided important first insights on how dam removals could affect N cycle processes in riparian zones and its implications for water quality and watershed management.

**Plain Language Summary** Dams are being removed to allow fish passage and improve safety for water users. Dam removal results in a drop of stream water level and a drying-out of the streamside (riparian) zones. We investigated if these changes would undermine the N-filtering service of riparian zones and increase N concentrations in groundwater and stream waters. We monitored soil and water N concentrations for 2 years following the removal of a 1.5 m milldam on Chiques Creek in Pennsylvania. Our data showed that while denitrification in soils did decrease, the N concentrations in riparian groundwaters and stream waters did not increase over the study period.

## 1. Introduction

Dams are increasingly being removed across the United States (US) (Bellmore et al., 2017; Foley et al., 2017). Since 1912, more than 1,490 dams have been removed across the US and Pennsylvania leads the nation in the number of milldams and their removals (American Rivers, 2020). Most (>90%) of these milldams are classified as low-head dams (height < 7 m) and are typically a relic of colonial and post-colonial era milling activities (Merritts et al., 2011; Walter & Merritts, 2008). Dam removal numbers could be higher since not all dam removals are recorded. This could particularly be true for the Mid-Atlantic Piedmont region, where thousands of small mill dams existed since the late 1600s (Walter & Merritts, 2008).

Low-head dam removals are primarily being driven by needs for public safety, reduction in financial liability, recreational access, aesthetics, and/or improvement in fish habitat (Bellmore et al., 2017; Foley et al., 2017;

Hart & Poff, 2002; Tonitto & Riha, 2016; Tullos et al., 2016). However, few follow-up, comprehensive studies are conducted on this large-scale, nationwide, experiment in reverse-engineering, and we know little about how this may impact waterways and landscapes. Much of the limited scientific research that has been done to date is focused on assessing how dam removals alter stream geomorphology, erode and transport sediments and nutrients or contaminants, and affect stream/aquatic habitat (Gold et al., 2016; Hart & Poff, 2002; Merritts et al., 2011, 2013; Miller et al., 2019; Pizzuto, 2002; Stanley & Doyle, 2002; Velinsky et al., 2006).

Dam removals can have strong influences on water quality by not only changing rates of sediment/nutrient and contaminant mobilization and transport, but also through the alteration of hydrologic and biogeochemical processes in the riparian zone. Dam removals, depending on the height of the dam, can cause significant drops in stream water level and this water level drop can extend for considerable distances upstream (Merritts et al., 2011, 2013). Because of the control that dam removals exert on the local water table, a removal can also result in decreased groundwater elevations in the riparian zone, upstream of the dam, which could lead to long-lasting ecological repercussions.

Riparian zones are critical to controlling the transport and mitigation of non-point nutrient pollution from uplands in many agricultural watersheds (Cole et al., 2020; Lowrance et al., 1997; Mayer et al., 2007; Peterjohn & Correll, 1984; Sweeney et al., 2004). The principal mechanisms for nitrate-N removal in riparian zones include denitrification of nitrate-N by anaerobic soil microbes and the assimilation/uptake of nitrate-N by riparian vegetation (Gold et al., 1998; Groffman et al., 1992; Hill, 2019; Lutz et al., 2020). These two processes are especially effective when groundwater levels in riparian zones are close to the soil surface and within the root zone (Lowrance et al., 1997). Riparian zones with elevated water tables are considered "hotspots" of denitrification and typically retain 50% – 90% of total nitrate-N loadings from upland sources (Gold et al., 2001; Lowrance et al., 1997; McClain et al., 2003; Vidon et al., 2010). Similarly, elevated water levels and riparian flooding associated with beaver dams has been shown to enhance nitrate-N loss via denitrification (Naiman et al., 1988). If the riparian zone groundwater elevations decrease following dam removals, it is possible that the ability of the riparian zone to denitrify or assimilate N through plant uptake could be diminished. Furthermore, drained riparian soils with deep groundwater levels could also result in soils actively mineralizing and nitrifying nitrogen (Appling et al., 2014; Gurwick, Groffman, et al., 2008, Gurwick, McCorkle, et al., 2008; Hill, 2011). This could result in nitrate-N leaching from the riparian zone and undermine its role as nutrient filter or buffer.

Despite the increasing rates of dam removal and its potential consequences for ecosystem services of riparian zones, the effects of dam removals on riparian N processing and filtering capacity have not been investigated. The only published work available is our recent perspective (Inamdar et al., 2021), which explores multiple hypotheses on how dam removals could potentially impact riparian zone processes and functions. Based on first principles, we (Inamdar et al., 2021) hypothesized that dam removals and dewatering of the riparian soils could result in reduced denitrification and plant uptake and an increase of nitrification in the riparian soils. This could result in an increase in nitrate-N leaching in ground and stream waters. Inamdar et al. (2021) however, also presented alternate hypotheses and mechanisms that could counteract the release and leakage of N from riparian zones post dam removal.

Here, we test some of the hypotheses presented in Inamdar et al. (2021) using data collected on riparian soils and water over 2 years following the removal of a small milldam (1.5 m tall) on July 9, 2018 on Chiques Creek in Pennsylvania (PA). Denitrification, nitrification, and mineralization rates, nitrate-N and ammonium-N concentrations, and stable isotopic content ($\delta^{15}$N) for riparian soils were determined. N concentrations for riparian soil water, groundwater, and stream water were also evaluated. Using these data, we addressed the key questions: How do denitrification and nitrification rates in riparian soil change with time following dam removal? How do ground, soil, and stream water nitrate-N concentrations evolve with time after dam removal? Does dam removal increase nitrate-N concentrations in riparian groundwaters after milldam removal? Our primary hypotheses (following Inamdar et al., 2021) were: H1: Following dam removal and drainage of riparian soils, denitrification rates will decline and nitrification rates will increase (Figure 1). H2: Riparian soil, ground and stream water nitrate-N concentrations will increase after dam removal indicating leakage of N from the riparian zone (Figure 1).

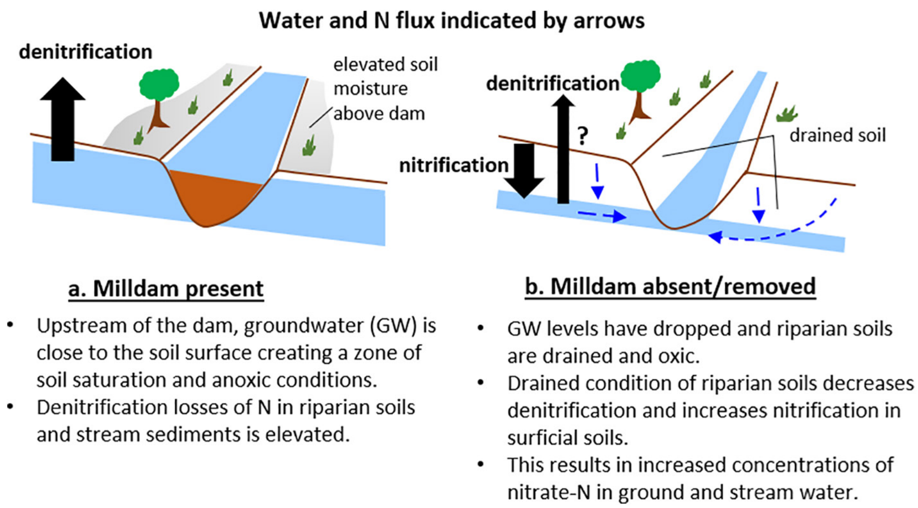


**Figure 1.** Conceptual model illustrating how groundwater level changes following dam removal affect N processes and leaching of nitrate-N in riparian ground and stream waters. The arrows represent the direction and magnitude of the N fluxes.

## 2. Site Description and Methods

### 2.1. Site Description

The Krady milldam was located on Chiques Creek in Rapho and West Hempfield Townships, Lancaster County, PA (40°04′08.2″N, 76°29′58.7″W), 5 km upstream of its confluence with the Susquehanna River, which drains into the Chesapeake Bay. The original dam was built in the 1700s for supplying water power to an adjacent grist and saw mill. Although the shape and size of the dam may have been modified since it was first built, it stood approximately 1.5 m tall and 30 m wide when it was removed on July 9, 2018. The catchment area draining to the dam is ∼159 km$^2$ and approximately 68% agricultural, 13% forested, 11% residential, and 7% grassland. The soils in the catchment and riparian zone are predominantly silt loams, but the riparian zone is also composed of fine-grained (silts and clays) legacy sediments (James, 2013) that have deposited upstream of the dam (Soil Survey, 2020). The thickness of the riparian sediments was ∼2.5 m and it contained buried organic horizons at various depths, particularly, closer to the stream edge. The geology of the upstream contributing drainage area is composed predominantly of dolomite/limestone (40%), shale (30%), and lesser degrees of arkosic sandstones, quartzites, conglomerates, phyllites and diabase intrusions (30%) (DCNR, 2020).

After the Krady milldam was removed in about 4–5 h, stream water levels at the dam dropped by about ∼ 1.5 m and resulted in exposure of large swaths of previously submerged riparian sediments (Figure 2). The drop in stream water surface progressively decreased and extended for about 1,755 meters upstream of the dam as determined from LIDAR differencing between 2019 and 2014 elevation surfaces for Pennsylvania. While stream water grab sampling (twice a week) was initiated on May 31, 2018, a little more than a month before dam removal, a comprehensive stream and riparian assessment was initiated four months after dam removal in November, 2018 (following the award of a National Science Foundation RAPID grant).

### 2.2. Hydrologic Monitoring for Stream Flow and Groundwater Elevations

One automated stream water level logger (U20L pressure transducer and logger from Onset Hobo Inc.) was installed in the creek bed just above the former dam location in November 2018 to monitor stream water levels every 30 min (Figure 3). Data from this water level logger along with stream velocity measurements using a Flow Tracker II and channel cross-sectional data were used to develop a stage-discharge relationship to compute streamflow discharge at the site.

Five groundwater wells (W1–W5, Figure 3) were augered down to ∼2 m on the riparian and legacy sediment terraces upstream of the dam over the period November 2018 through April 2019 (Figure 3). Two additional

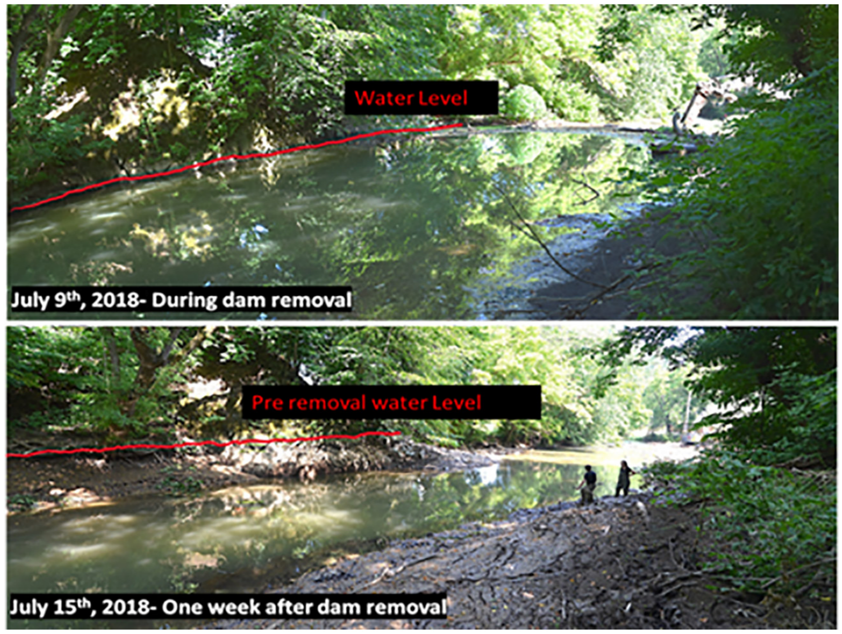


**Figure 2.** Chiques Creek in Pennsylvania during (top) and after removal (bottom) of the Krady dam. Stream and adjacent groundwater levels dropped by about 1.5 m following dam removal. View is looking downstream towards the dam. The tractor dismantling the dam can be seen on the right in the top panel and some drainage of stream water had already commenced in the top panel.

wells (W1b and W3b adjacent to W1 and W3, respectively) were added in November 2019 to a greater depth of ∼3 m since the existing wells and their sensors were drying out over the summer. The wells were made of 5 cm PVC pipes that were screened below the soil surface. The wells were located where site access and permissions were available. The wells were equipped with Hobo U20L water level loggers to measure groundwater levels every 30 min. The measured stream and groundwater levels were georeferenced to soil surface elevations based on a real-time kinematic GPS survey. The groundwater elevations were used to monitor riparian drainage post dam removal and the frequency of wet-dry cycles experienced by the riparian soils over the study period. Since well W2 dried up early in the study, data from that well is not included.

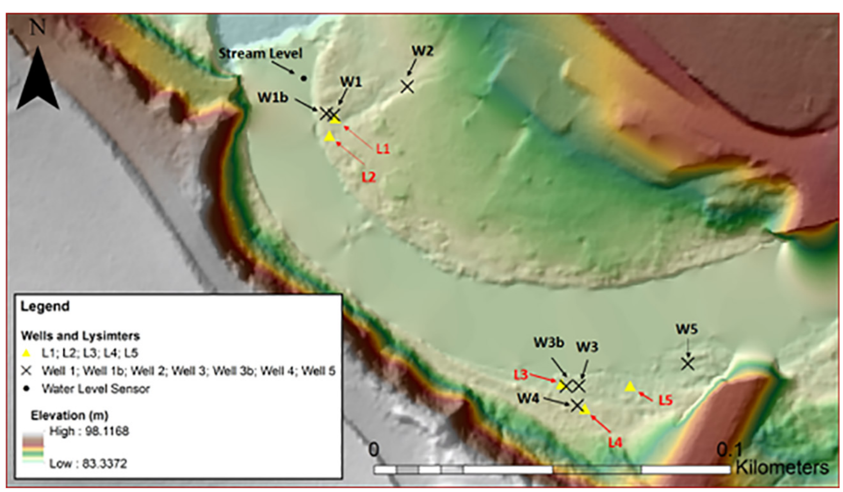


**Figure 3.** Locations of stream water level sensor, groundwater wells (W), and soil water lysimeters (L) upstream of the former Krady dam. The LIDAR digital elevation model (DEM) image was taken when the dam was present and indicates the pre-dam stream water level.

### 2.3. Water Quality Sampling and Analysis

Water samples from the stream and the groundwater wells were collected every two weeks over 2018–2019 and monthly in 2020 (due to Covid-19 travel and access constraints). Stream water sampling was initiated on May 31, 2018 while riparian groundwater sampling was initiated between December 2018 and January 2019 for wells W1–W5 and November 2019 for wells W1b and W3b. Groundwater samples were collected using a battery powered peristaltic pump in clean, 250 mL polythene bottles. Stream water samples were collected manually immediately below the former dam location (Figure 3).

To characterize soil water chemistry for the near-surface horizon (top 30 cm), five shallow soil lysimeters were installed in the riparian soils in October of 2019. All lysimeters were installed within close proximity of the groundwater wells (Figure 3). The lysimeter (1900L series, Soil Moisture Inc.) was a PVC pipe 5 cm in diameter with a suction cup at the bottom. Each lysimeter was buried in the upper 20–30 cm of the riparian soil at an angle of 45° and repacked with the same augered soil. Using a portable hand pump, each lysimeter chamber was pressurized to ∼340 kPa (50 psi) prior to sampling. Water samples were collected using a standard plastic syringe connected to a short length of tubing. Soil water sampling was performed on a biweekly basis starting November 2019 until February 2020 (terminated thereafter due to Covid-19 restrictions).

No sampling was performed between February and May 2020 due to lockdowns associated with Covid-19. All water samples were placed on ice after collection and filtered in the laboratory with a 0.7 micron GFF. These samples were analyzed for total dissolved organic carbon (DOC), total dissolved N (TDN), nitrate-N, and ammonium N at the University of Delaware Soils Laboratory. Nitrate-N and ammonium-N were measured colorimetrically using a Bran & Luebbe Autoanalyzer 3 (Bran & Luebbe). TDN and non-purgeable DOC were measured by combustion using an Elementar Vario-Cube TOC Analyzer.

### 2.4. Riparian Soil Sampling and Analysis

Riparian soil sampling was performed to assess the changes in N concentrations (nitrate-N, ammonium-N, and total N) and N processes (denitrification, nitrification, and mineralization) following dam removal. Selected soil subsamples were also analyzed for stable soil nitrogen isotopes ($\delta^{15}$N) to provide insights into isotopic changes and the potential role of denitrification and nitrification in shaping the isotopic values. Denitrification has been shown to enrich or increase the $\delta^{15}$N values for soils whereas lack of denitrification will result in depleted $\delta^{15}$N values (Evans, 2007; Kendall et al., 2007). Soil samples were collected by manually augering the riparian soils upstream of the dam at multiple depths near (within 2 m) the groundwater wells. Some of these samples were obtained during the augering of the wells. Soils samples for N concentrations and process rates were available for five dates over the 2018–2020 period (November 2018, April, August, and November 2019 and July 2020). Soils samples for isotopic analysis were available for all except the November 2018 date. A planned soil sampling date for April 2020 was missed because of Covid-19 constraints. All soil samples were collected in ziplock bags and stored on ice in the field. Soil samples were analyzed for $\delta^{15}$N at the University of Maryland Center for Environmental Science isotope facility using a Thermo Delta V Ratio Mass Spectrometer (Thermo) interfaced with an elemental combustion system (4010 CHNSO analyzer, Costech). Soils samples for nitrate-N and ammonium-N were extracted using KCl and the extract analyzed colorimetrically (as described above).

Denitrification enzyme assay (DEA), nitrification, and mineralization analyses were performed at the University of Rhode Island Watershed Hydrology Laboratory. Since DEA assays are conducted in ideal laboratory conditions they typically provide the maximum potential denitrification rates as opposed to in-situ values (Groffman et al., 1993, 2005). These assays have been valuable in situations where comparisons of denitrification changes are to be assessed across space and time (Groffman et al., 2005). DEA samples were processed for unamended conditions as well as amended (labile C) conditions. DEA analyses were performed by homogenizing the soil sample with DI water and sodium nitrate ($NaNO_3$), purging the sample container with helium and acetylene gas in order to induce anoxia and to prevent the conversion of $N_2O$ to $N_2$ gas. In the case of amended DEA assays, glucose was added to prevent soil enzyme activity from being carbon limited. Headspace gas was then sampled from the sample container and analyzed using gas chromatography in order to quantify the denitrification product ($N_2O$) gas in μg kg$^{-1}$ h$^{-1}$.

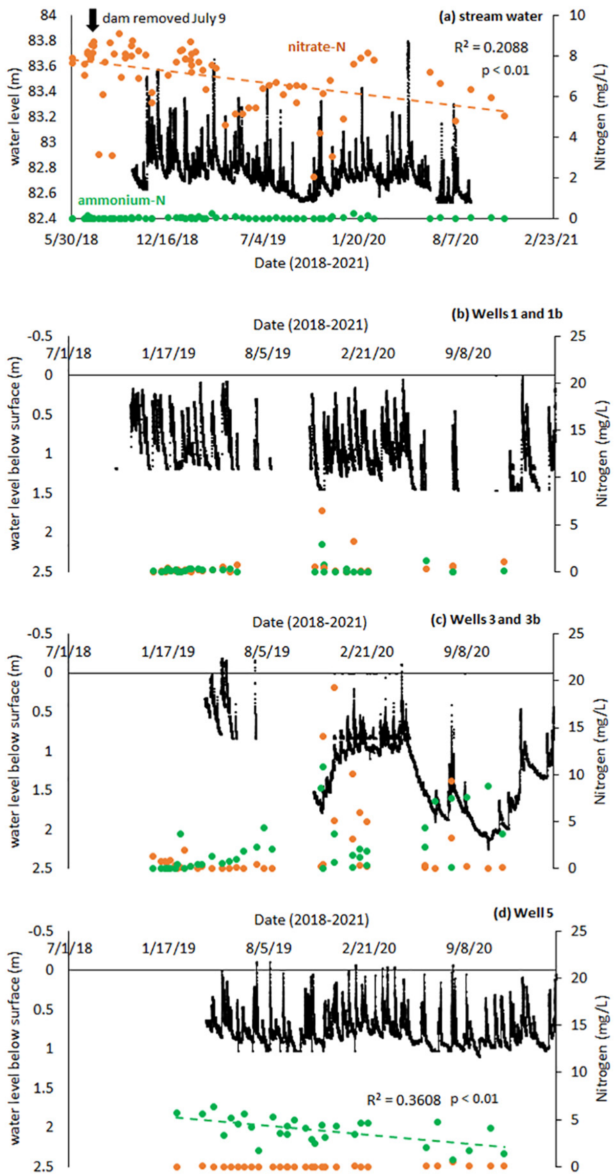


**Figure 4.** Time series of stream water level and groundwater levels (black lines) below the soil surface (m) and concentrations of nitrate-N (brown circles) and ammonium-N (green circles) (mg N $L^{-1}$).

Net nitrification and mineralization incubations were performed using a week-long incubation in the laboratory. Soils were incubated at 25°C and a measured amount of water was added to maintain the starting soil/sample weight. The difference in ammonium-N and nitrate-N concentrations pre- and post-incubation provided the net ammonification and nitrification rates, respectively, and all results were reported in $\mu g\ kg^{-1}\ h^{-1}$. N process rate comparisons were performed for the 0–1 m soil profile and greater than 1 m depth.

### 2.5. Data Synthesis and Analysis

Water and soil data were analyzed to assess N trends with time (since dam removal) and soil depth. Time series of stream and groundwater levels and N and DOC concentrations were plotted, and any significant temporal trends were determined using Pearson regression. Changes in time for soil N and C concentrations, N process rates, and $\delta^{15}N$ values was assessed by pooling the data for the sampled dates, presenting them in box and whisker plots, and determining significant differences among the dates using Student's t test (α level of 0.05). All statistical analysis was performed using SAS-JMP software.

## 3. Results

### 3.1. Stream Water Levels, Flow, and Groundwater Elevations

Average stream water level in Chiques Creek was 82.8 m above sea level over the duration of the study (range: 82.5–83.8 m; Figures 4a and S1). The average streamflow discharge for the same period was 2.11 $m^3/s$ with a maximum of 15 $m^3/s$ on May 1, 2020 following a large storm. Stream baseflow was highest during winter and spring and lowest in late summer and early autumn (September-October).

Riparian groundwater elevations were generally greater than stream water levels (Figure S1) and during the wettest periods the level of the groundwater was about 1 m above the stream water level. However, during the driest periods in August–September, when evapotranspiration demands were high, water levels for wells W1–W4 occasionally fell below the stream water level. With respect to the soil surface (Figures 4b–4d and S2), riparian water levels fluctuated from surficial ponding (negative values) to water levels more than 2 m below the surface (except W5). Levels for wells W1–W4 also dropped below the level of the sensors and thus no water level was recorded (indicated by gaps in Figure 4). Wells W5 (closest to the stream and wet year-round) and W3b provided the most continuous water level data for the study period.

Other than the sharp drop in levels (~1.5 m) immediately after dam removal, stream and groundwater levels did not show any additional long-term increasing or decreasing trends (since start of water level monitoring in November 2018). It should be noted though that 2018, the year of dam removal, was one of the wettest on record for the region with 1,631 mm of precipitation recorded at the Lancaster Airport, approximately 20 km from the study site (Pennsylvania State Climatologist, 2021). Corresponding annual precipitation amounts for 2019 and 2020 for Lancaster Airport were lower at 1,119 mm and 1,080 mm, respectively. The average annual precipitation for Lancaster area is 1,066 mm. There were a number of large precipitation events (Figure S3a) that occurred soon after dam removal with peak flows and significant flooding on July 25 and August 4, 2018 (Figure S3b; data recorded at the Susquehanna River Basin Commission (SRBC) Chiques Creek gaging station 6.4 kilometers upstream of this dam removal site).

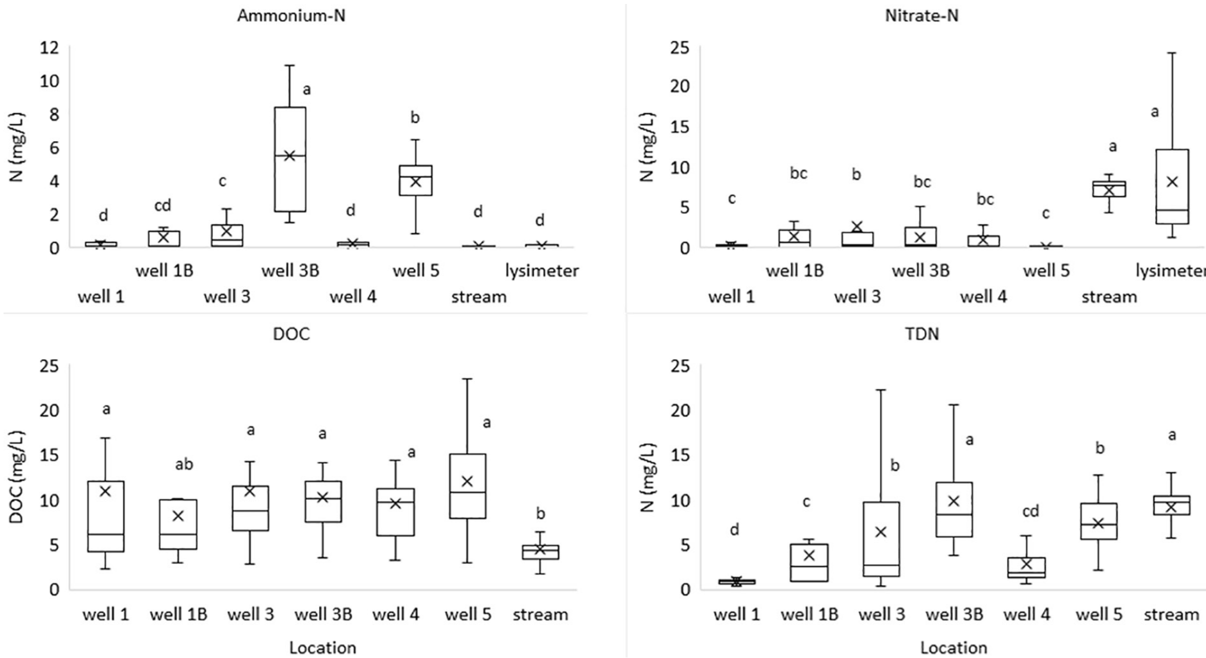


**Figure 5.** Box and whisker plots comparing concentrations for dissolved nitrate-N, ammonium-N, dissolved organic carbon (DOC) and total dissolved N (TDN) in individual groundwater wells, stream water and all soil water lysimeters. Data for lysimeters was not available for DOC and TDN. Locations with different letters are significantly different at an α level of 0.05. The lower bound of the box represents the first quartile (25%), the upper bound represents the third quartile (75%), the horizontal line in the box is the median, the "x" mark is the mean, and the whiskers indicate the minimum and maximum values. Number of samples collected for each location over the study period indicated in brackets: well 1 (25), well 1b (9), well 3 (28), well 3b (12), well 4 (17), well 5 (29), stream (83), and lysimeter (29).

### 3.2. Nitrogen Concentrations in Stream, Ground, and Soil Water

Average stream water nitrate-N concentrations for Chiques Creek were 7.05 mg N $L^{-1}$ with a minimum and maximum of 2.06 and 9.10 mg N $L^{-1}$, respectively (Figure 5). Stream water nitrate-N concentrations were particularly high during the summer and fall of 2018 (Figure 4a), likely in response to the multiple storms and wet hydrologic conditions during that period. Storm related increases were also observed in spring 2019 and 2020. Overall, a significant (Pearson $R^2 = 0.208$, $p < 0.01$) declining trend in stream water nitrate-N concentrations is observed (Figure 4a). In contrast to nitrate-N, stream water ammonium-N concentrations were low and close to detection levels (Figures 4a and 5).

In contrast to stream water, nitrate-N concentrations in riparian groundwater were generally low and variable (Figures 4 and 5). Average nitrate-N concentrations for wells were: W1 = 0.18 mg N $L^{-1}$, W1b = 1.72 mg N $L^{-1}$, W3 = 2.43 mg N $L^{-1}$, W4 = 0.72 mg N $L^{-1}$, W5 = 0.02 mg N $L^{-1}$ (Figure 5). In comparison to nitrate-N, average ammonium-N concentrations were below 1 mg/L for all groundwater wells other than wells W3b (4.75 mg N $L^{-1}$) and W5 (4.27 mg N $L^{-1}$) (Figure 5). Ammonium-N response for near-stream well W5 was unique from the other riparian wells (Figure 4d), with elevated concentrations in the range of 0.8–5 mg/L and a consistent and significant ($R^2 = 0.36$, $p < 0.01$) decline in concentrations since the start of sampling in February 2019. Groundwater concentrations for both nitrate-N and ammonium-N occasionally increased sharply during storms following an extended dry period–as seen for wells W3 and W3b in the autumn of 2019 and the summer of 2020 (Figures 4b and 4c). Nitrate-N concentrations spiked up to 19 mg N $L^{-1}$ while ammonium-N concentrations exceeded 10 mg N $L^{-1}$ for wells W3 and W3b (Figures 4b and 4c).

TDN varied considerably from 1 to 30 mg N $L^{-1}$ in both stream and groundwater (Figures 5 and 6). For stream water, on average, nitrate-N composed 77% of TDN. The significant decline observed for nitrate-N in stream water was also observed for TDN with concentrations decreasing significantly ($R^2 = 0.22$, $p < 0.01$) over the study period (Figure 6a). For riparian groundwater wells, other than W5, there were no consistent long-term trends in TDN. For W5, TDN concentrations (Figure 6d) declined significantly ($R^2 = 0.18$,

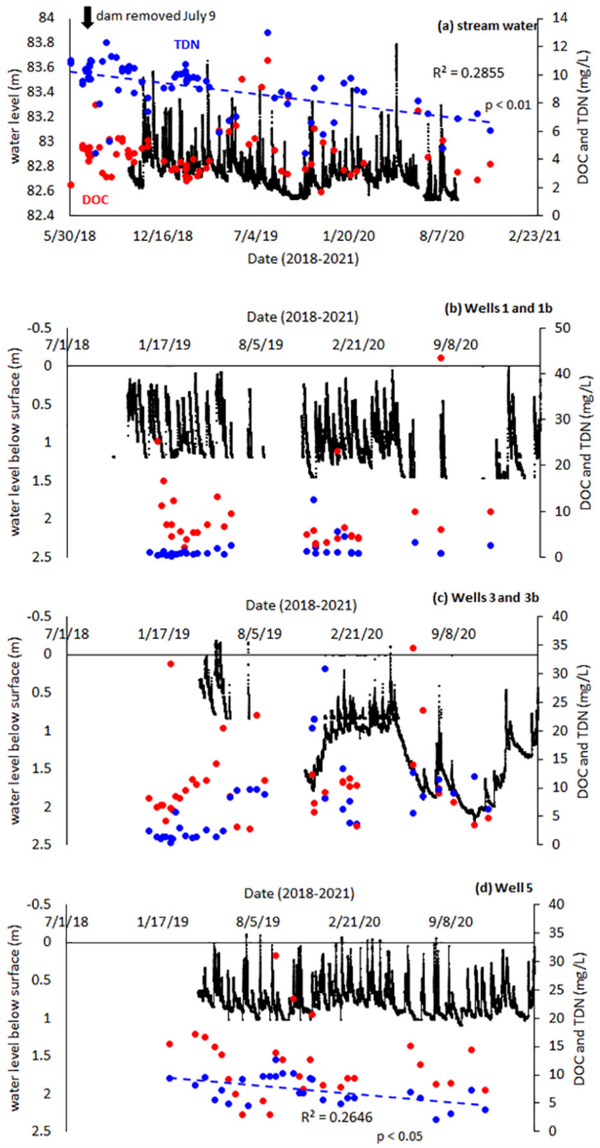


**Figure 6.** Time series of stream water level and groundwater levels (black lines) below the soil surface (m) and concentrations of total dissolved N (blue circles, mg N/L) and dissolved organic carbon (red circles, mg C/L).

$p < 0.05$), mirroring the decline in ammonium-N, which, on average, constituted 54% of the TDN. Similar to TDN, DOC displayed high variability (Figure 6), however, concentrations of DOC were consistently found to be greater in groundwater well samples (>0–50 mg C $L^{-1}$) than in stream water (>0–15 mg C $L^{-1}$). No long-term increasing or decreasing trends were observed for DOC since the start of the monitoring.

All soil water lysimeters had higher concentrations of nitrate-N compared to the groundwater wells (Figure 5). For the same length of time (November 2019–February 2020) during which lysimeters were measured, the average concentration of nitrate-N measured in soil water was 7.38 mg N $L^{-1}$ versus 1.35 mg N $L^{-1}$ measured in the groundwater. The average ammonium-N measured for all lysimeters was low ranging from 0.03 to 0.13 mg N $L^{-1}$.

### 3.3. Soil Nitrate-N and Ammonium-N Concentrations

Soil nitrate-N concentrations varied between 0 and 25 mg $kg^{-1}$ with higher values for surficial soils (0–1 m) and a sharp decline in concentrations with soil depth (Figures 7 and S4). The only significant ($p < 0.05$) nitrate-N change was the sharp drop in soil nitrate-N concentrations between November 2019 and July 2020 for the top 1 m soil depth (Figure 7). In contrast to nitrate-N, soil ammonium-N concentrations were more variable with values spanning 0–240 mg $kg^{-1}$. Contrary to the depth pattern of nitrate-N, ammonium-N concentrations were low near the soil surface and increased sharply beyond a soil depth of 1–1.5 m (Figure S4). Ammonium-N concentrations for the deep soils (>1 m depth) decreased significantly ($p < 0.05$) in July 2020 (Figure 7).

### 3.4. Denitrification, Nitrification, and Mineralization of Riparian Soils

The largest and most significant changes in N process rates occurred for the 0–1 m depth (Figure 8). As expected, amended denitrification rates were greater than the corresponding unamended values because of labile C addition (Figure 8). For the top 1 m, a large and significant ($p < 0.05$) decrease in amended denitrification was observed between April and August 2019, but there were no significant changes thereafter. On the other hand, unamended denitrification values (0–1 m) decreased earlier with a significant ($p < 0.05$) drop between November 2018 and April 2019. Nitrification values (0–1 m) were generally lower than the corresponding denitrification rates and there was no statistically significant change with time (Figure 8). Similarly, mineralization rates for the 0–1 m depth were low and did not yield any significant changes with time. The denitrification rates for the deeper soils (>1 m) were low and did not change significantly other than the decrease in unamended values between November 2019 and July 2020. Nitrification and mineralization values for the deeper soils (>1 m) were greater than those for 0–1 m depth, but did not reveal any significant changes with time (Figure 8).

### 3.5. Soil $\delta^{15}N$ With Depth and Time After Dam Removal

Riparian soil $\delta^{15}N$ values were highest for April 2019 (0–1 m depth = 10.9 ‰ and >1 m depth = 9.6 ‰; Figure 9). These values decreased sharply and significantly ($p < 0.05$) by August 2019 and no significant changes were observed thereafter. The decrease or depletion of $\delta^{15}N$ individual values was particularly pronounced for near-surface soil samples (Figure S5).

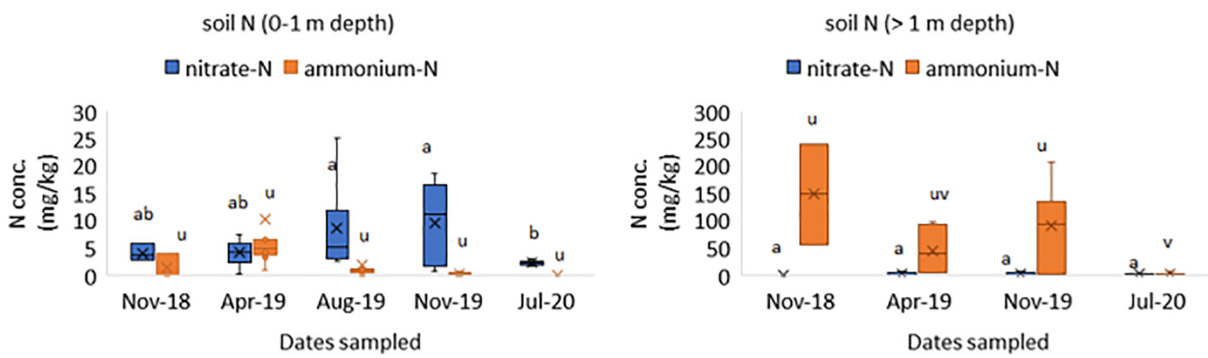


**Figure 7.** Soil concentrations of nitrate-N and ammonium-N (mg $kg^{-1}$) for 0–1 m and >1 m depths for the five sampled dates across 2018–20. Dates are expressed in month and year. Statistical comparisons are performed for nitrate-N (indicated by letters a and b) and ammonium-N (letters u and v) for the dates sampled. Dates with different letters are significantly different at an $\alpha$ level of 0.05. The lower bound of the box represents the first quartile (25%), the upper bound represents the third quartile (75%), the horizontal line in the box is the median, the "x" mark is the mean, and the whiskers indicate the minimum and maximum values. Sample numbers, in brackets, for 0–1 m depth were: November 2018 (3), April 2019 (13), August 2019 (10), November 2019 (5), and July 2020 (6). Sample numbers for >1 m depth were: November 2018 (2), April 2019 (4), November 2019 (9), and July 2020 (6).

## 4. Discussion

The suite of water and soil data presented above clearly shows that important changes occurred in riparian soils and groundwaters during the 2 years following dam removal. Some of these changes support our hypotheses, while others do not. In addition to nitrate-N, the soil and groundwater data also revealed elevated ammonium-N concentrations and a significant decline in those concentrations with time. Our results also demonstrate that the rate, extent, and timing of N changes differed between groundwaters and soils and also among N processes in the riparian soils. We elaborate on these changes in light of the hypotheses, present an updated conceptual model considering ammonium-N, discuss key caveats, and assess the broader implications of these results.

### 4.1. Changes in Denitrification, Nitrification, and Mineralization in Riparian Soils Following Dam Removal

Our primary hypothesis H1 (Figure 1) was that riparian soil drainage and aeration following dam removal would decrease denitrification rates and simultaneously increase nitrification potentials in riparian soils. This was based on well-established studies indicating that denitrification is enhanced under saturated and anoxic soil conditions associated with near-surface groundwater levels while nitrification is favored under well-drained and oxic soil conditions (Burt & Pinay, 2005; Cirmo & McDonnell, 1997; Gold et al., 1998). This assessment was also supported by work of Weitzman and Kaye (2017) who reported elevated rates of nitrification in surficial and well-drained riparian sediments and suggested these soils could serve as potential nitrate-N sources for streams. The nitrification and N leaching rates could especially be elevated if riparian sediments contain buried organic-rich soil layers which can be mineralized following drainage and oxidation (e.g., Gurwick, Groffman, et al., 2008, Gurwick, McCorkle, et al., 2008; Hill, 2011). Riparian sediments upstream of milldams have been found to contain such buried organic horizons with carbon contents in the range of <1% to greater than 7% (Lutgen et al., 2020; Sienkiewicz et al., 2020). We also noted buried organic matter (e.g., stacks of dark, decomposing leaf layers and wood) while augering for groundwater wells, particularly near the stream. Occasional dissolved oxygen measurements for near-stream wells (especially W5) also indicated anoxic (<0.5 mg/L) groundwater conditions.

The N process rates produced mixed results in support of H1. Amended and unamended denitrification clearly indicated a drop in denitrification (Figure 8) for riparian soils although the timing of the decrease (November 2018 vs. August 2019) differed between the two methods. Particularly striking was that both approaches supported a significant downward shift in denitrification early on in the study. This was despite the fact that sampling was performed across different seasons with varying soil moisture, temperature, and other seasonal variables which could have influenced the results (even though the actual assays were conducted in controlled laboratory conditions, the starting soil sample moisture and temperature conditions

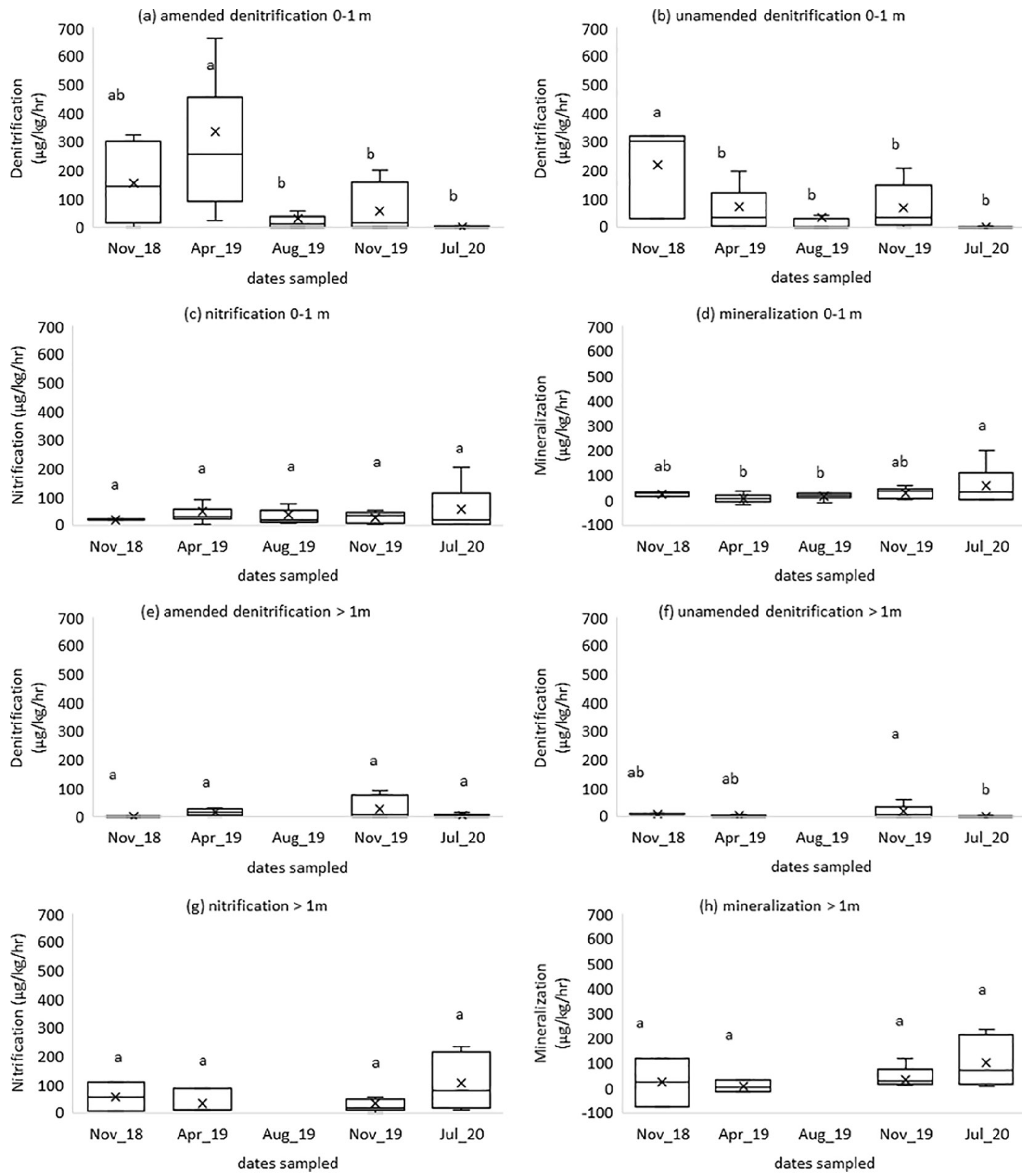


**Figure 8.** Soil N process rates for various sampled dates pooled for depths 0–1 m and >1 m. Dates with different letters are significantly different at an α level of 0.05. The lower bound of the box represents the first quartile (25%), the upper bound represents the third quartile (75%), the horizontal line in the box is the median, the "x" mark is the mean, and the whiskers indicate the minimum and maximum values. Sample numbers, in brackets, for 0–1 m depth were: November 2018 (3), April 2019 (13), August 2019 (10), November 2019 (5), and July 2020 (6). Sample numbers for >1 m depth were: November 2018 (2), April 2019 (4), November 2019 (9), and July 2020 (6).

varied seasonally). Contrary to denitrification, the nitrification and mineralization assays, indicated no change in rates and thus did not support hypothesis H1. The magnitude of nitrification and mineralization in the top 1 m of the soil, at least for the early sampling dates, were lower than the denitrification rates, suggesting that while denitrification was decreasing with time, it was still a dominant N process in the riparian topsoil.

Following H1, if denitrification is declining and nitrification is increasing in the drained riparian soils, one would expect soil nitrate-N concentrations to increase. While our data suggests some accumulation in the top 1 m of the riparian soil (Figure 7), the increase was not significant. Thus, similar to nitrification rates,

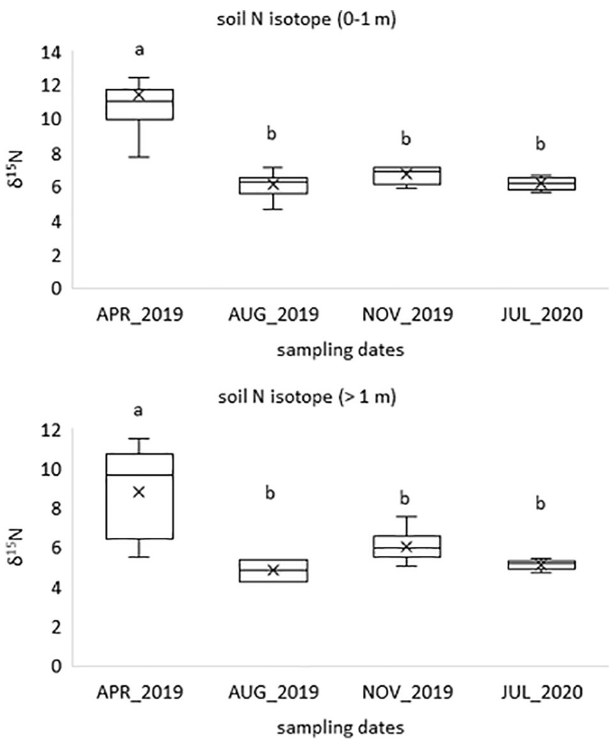


**Figure 9.** Soil $\delta^{15}$N values for sampled dates following dam removal pooled for soil depths 0–1 m and >1 m. Dates with different letters are significantly different at an α level of 0.05. The lower bound of the box represents the first quartile (25%), the upper bound represents the third quartile (75%), the horizontal line in the box is the median, the "x" mark is the mean, and the whiskers indicate the minimum and maximum values. Sample numbers, in brackets, for 0–1 m depth were: April 2019 (12), August 2019 (11), November 2019 (4), and July 2020 (9). Sample numbers for >1 m depth were: April 2019 (5), August 2019 (2), November 2019 (14), and July 2020 (9).

data on soil nitrate-N concentrations did not conclusively support hypothesis H1. While ammonium-N values were low in the top 1 m and did not change with time, they were more than an order of magnitude greater in the subsurface soils (>1 m depth). These elevated ammonium-N values for deeper soils could be due to suppression of ammonium consumption by nitrification (e.g., Hefting et al., 2004; Hill & Duval, 2009) and/or dissimilatory nitrate reduction to ammonium (DNRA, Pandey et al, 2020; Rütting et al, 2011; Sgouridis et al., 2011), both of which typically occur under reducing and anoxic soil conditions.

If drainage associated with milldam removal causes riparian soils to become oxic, one would expect that the soil ammonium-N concentrations would decline. While our soil ammonium-N data indicates an insignificant decline initially (Figure 7), we do see a significant drop in soil ammonium-N concentrations between November 2019 and July 2020. This decrease would suggest that drier soil conditions and drainage following dam removal are depleting the ammonium-N pool that had accumulated in the deeper riparian soils (>1 m). This depletion or loss of soil ammonium could have occurred through nitrification or desorption of ammonium by drainage waters. The elevated values of soil ammonium-N that were measured for the deeper soils were likely responsible for the elevated nitrification rates that were measured in the laboratory assays for the deep (>1 m) soils (Figure 8).

The strongest and most significant early shift in N status of the riparian soils was indicated by the soil $\delta^{15}$N values (Figure 9). Unlike N process rates and concentrations, which provide a discrete assessment of N status in time, $\delta^{15}$N values provide a cumulative, time-integrated signature of multiple N processes, particularly denitrification and nitrification (Evans, 2007; Kendall et al., 2007). Reducing soils with elevated denitrification conditions increase or enrich $\delta^{15}$N values, while oxic soils with low denitrification typically produce soils with depleted or low $\delta^{15}$N values (Evans, 2007; Kendall et al., 2007). Thus the $\delta^{15}$N value measured for soils reflects the net effect of these diverse processes and can be considered a more robust metric of N change. The significant decrease or depletion in $\delta^{15}$N for both 0–1 m and >1 m soil depths in August 2019 (Figure 9) suggests a decrease in denitrification processing of N in riparian soils and/or a drainage loss of enriched nitrate-N. The $\delta^{15}$N results support hypothesis H1 and also agree with the changes in amended denitrification for 0–1 m (Figure 8). The sharp and early decrease in soil $\delta^{15}$N values was unexpected, given that, typically, soil $\delta^{15}$N values are expected to change slowly (Evans, 2007). The unusually high rainfall and hydrologic activity in the summer and autumn of 2018, post dam removal, could have contributed to this early shift in $\delta^{15}$N values (elaborated further in the section below). This also suggests that soil $\delta^{15}$N could be a good, sensitive, metric of N change in soils.

### 4.2. Soil, Ground, and Stream Water Nitrate-N Concentrations Following Dam Removal

Hypothesis H2, a follow-up to H1, was that if nitrification exceeded denitrification in soils, elevated nitrate-N concentrations would be observed in riparian soil, ground, and stream waters and that these concentrations would progressively increase with time after dam removal. Similar to H1, observations of dissolved N in soil, ground, and stream water presented a complex picture with mixed results. Stream water nitrate-N concentrations were indeed elevated in the summer and autumn of 2018 after the July dam removal. The elevated concentrations were likely associated with hydrologic flushing of N (Burns, 2005) from watershed soils following the large storms and unusual levels of rainfall in the region. Such storm-associated N increases have been reported across various land uses, and the magnitude of the increase is a function of the amount of nitrate-N available in surficial soils and the intensity and magnitude of the storms (Inamdar et al., 2006;

Oeurng et al., 2010; Vaughan et al., 2017). Thereafter, however, nitrate-N concentrations for stream water showed a significant and continuous decline over the next 2 years. The declining trend was contrary to the hypothesized N leakage and increase in nitrate-N as per H2. This long-term decline in stream water nitrate-N has also been observed at the SRBC gaging station 6.4 km upstream and in other tributaries of the Chiques Creek (SRBC, personal communication). Thus, the Chiques Creek nitrate-N decline is a watershed wide trend and could be associated with nutrient conditions returning to more "normal" lower concentrations following a very wet 2018. Irrespective of the specific drivers of N decline, it is clear that the stream water N decline is not influenced by changes related to a single dam removal and thus our stream water N data do not support hypothesis H2. Dam removal may increase watershed flux of N, but this is thought to occur where large reservoirs (relative to the contributing watershed) are drained (Gold et al., 2016).

Surficial soil water nitrate-N concentrations recorded by soil lysimeters were highest of all riparian sampling locations suggesting that mineralization and nitrification of N were likely responsible for the elevated values. In comparison to soil water, nitrate-N concentrations in riparian groundwaters were very low. The few instances when groundwater nitrate-N concentrations spiked were during rewetting following a dry period (e.g., W3b during rewetting in late 2019, Figure 4). This suggests that while nitrate-N was indeed being produced in surficial soil water and recharging the groundwaters during stormflows, nitrate-N concentrations did not persist for long in groundwaters. Thus, while soil water nitrate-N concentrations supported H2 (nitrate-N release following dewatering of riparian soils), groundwater N data did not support this hypothesis. It appears that nitrate-N removal mechanisms continue to persist in riparian groundwaters despite a 1.5 m drop in groundwater levels and substantial loss of reducing/anaerobic soil volume. One possibility could be that denitrification "hotspots" (e.g., Vidon et al., 2010) continue to persist at the groundwater interface and are responsible for the removal of nitrate-N. It is also likely that a more "dynamic" and variable groundwater regime following dam removal is enhancing denitrification losses of N (Inamdar et al., 2021). Previous research has shown that stagnant moisture regimes, that are continuously wet or dry, can depress denitrification process rates (Bernard-Jannin et al., 2017; Guo et al., 2014; Shi et al., 2020; Tomasek et al., 2019; Ye et al., 2017). Conversely, large hydrologic variations or drying-wetting soil moisture cycles have been shown to prime and increase denitrification through fresh inputs of C and N associated with mineralization and nitrification (Shi et al., 2020; Ye et al., 2017). If this happens, the loss of anaerobic denitrification soil volume could be offset by increasing soil denitrification rates driven by a more dynamic groundwater and soil moisture regime (Inamdar et al., 2021).

Another possibility that could explain the low concentrations of nitrate-N in groundwaters and the large disparity with overlying soil water nitrate-N is the occurrence of DNRA (Burgin & Hamilton, 2007; Pandey et al., 2020; Rütting et al, 2011; Sgouridis et al., 2011) in riparian groundwaters. DNRA is an anoxic soil process that converts nitrate-N to ammonium-N and competes with denitrification (Pandey et al., 2020). DNRA is favored over denitrification under elevated DOC to nitrate-N ratios (>12; Pandey et al., 2020; Rütting et al, 2011; Sgouridis et al., 2011, Wang et al., 2020). In addition, fine soil particles such as clays and silts, that are abundant in milldam legacy sediments, have been reported to enhance DNRA through their influence on water filled pore space and redox potentials (Chen et al., 2015; Sgouridis et al., 2011). Occurrence of DNRA would also explain the elevated ammonium-N concentrations observed in groundwaters and the deeper soil profiles. Well W5, in particular, displayed elevated dissolved ammonium-N values in groundwaters which declined steadily and significantly over the study period (Figure 4d). DOC concentrations in this well were also high (Figures 5 and 6d) with a mean DOC to nitrate-N concentration (mg $L^{-1}$) ratio of 840 and range of 16–3,100; elevated values that would potentially favor DNRA over denitrification.

We speculate that deep (>1 m) riparian soils and particularly those adjacent to the stream with anoxic environment and buried organic horizons, provide ideal conditions for DNRA to occur and contribute to the elevated ammonium-N values we recorded at W5. We suspect that occurrence of DNRA and accumulation of ammonium in groundwaters has also likely contributed to the elevated soil ammonium-N values (Figure 7) through sorption of dissolved groundwater ammonium-N on fine-grained legacy sediments. This accumulated ammonium-N in near-stream, DOC-rich, riparian groundwaters and legacy sediments is likely slowly released over time–as indicated by the declining trend in groundwater ammonium-N for W5 (Figure 4d). It should be noted though that despite the elevated ammonium-N in W5 groundwaters, only a few meters from the stream, stream water concentrations of ammonium-N were very low or below detection.

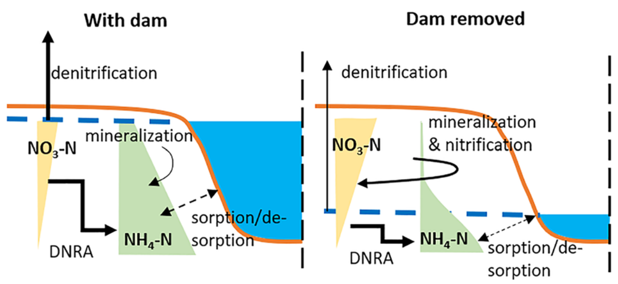


**Figure 10.** Updated conceptual model that highlights the role of ammonium-N pool (green) alongside the nitrate-N pool (yellow) in the presence of the dam (left) and after dam removal (right). The potential contribution of dissimilatory nitrate reduction to ammonium to the nitrate-N consumption and ammonium-N production is also included. The arrows indicate the direction and magnitude (thickness of arrow) of the N fluxes.

It is possible that release of ammonium-N into stream waters is likely consumed rapidly or converted to nitrate-N in the more oxic environment of the stream. Recent work of Zhao et al. (2021), for cascade dam reservoirs in China, suggest that anoxic conditions associated with small dams could increase the sediment pore water concentrations of ammonium-N (at the cost of nitrate-N) and the accumulated sediment pore water ammonium could diffuse into overlying waters posing a water quality risk.

### 4.3. Updated Model of N Changes in Riparian Soils Following Dam Removal

In our hypotheses and conceptual model (Figure 1), we emphasized the role of nitrate-N in driving the changes in N conditions following dam removal. Our observations from this study, however, suggest that ammonium-N could also play a significant role in retention and release of N in such riparian settings. Similarly, organic N associated with the buried organic material could also be an important factor. Thus, we provide a revised conceptual model (Figure 10) with the potential of nitrate-N and ammonium-N as dual inorganic N reservoirs with variable release rates influencing N status in milldam riparian sediments and groundwaters.

The accumulation of ammonium-N in anoxic environments like wetlands (e.g., Jahangir et al., 2017; Rahman et al., 2019; and Zhao et al., 2021) and stream-riparian margin sediments (Duval & Hill, 2007) has been reported before. Jahangir et al. (2017) and Zhao et al. (2021) attributed the ammonium accumulation to DNRA, while Duval and Hill (2007) attributed it to anaerobic mineralization of buried organic material and suppression of nitrification. We propose that in settings similar to those found in the Piedmont, anthropogenic legacies associated with milldams (i.e., deposition of fine-grained silt and clays and burial of fluvial organic matter upstream of the dams), can enhance ammonium-N production and accumulation in riparian soils. We suspect that anoxic and C-rich conditions increase the pool of ammonium-N and regulate the release or loss of nitrate-N through processes like DNRA and suppression of nitrification. As proposed in the original conceptual model, nitrate-N release from dewatered, oxic, surficial sediments does occur and likely contributes to the early pulse of N exports, especially during storms. Any additional nitrate-N is likely denitrified in the subsoils and groundwaters and/or is converted to ammonium-N via DNRA, particularly along the anoxic, C-rich, near stream riparian boundary. A portion of the dissolved ammonium-N is sorbed on fine sediments and provides an additional N reservoir. Thus, this fine-grained, C-rich riparian boundary, could be serving as a "ammonium-N fringe or reservoir" adjacent to the stream in conserving N as ammonium (as opposed to its loss via denitrification). These dissolved and sorbed ammonium-N stores could slowly release N over a longer period, post drainage, through processes like desorption, anaerobic ammonia oxidation (anammox, Gao et al., 2018), nitrification, and denitrification. The amount of ammonium-N stored and the rate and time required to exhaust the ammonium-N pool in the near-stream region (post dam removal) could have important ecosystem and environmental implications.

### 4.4. Caveats and Additional Considerations

While this study provided novel and valuable insights into N changes in riparian soils following dam removal, there are important aspects that should be considered that can help strengthen future studies. We were unable to collect any pre-dam removal data at this site because of lack of advanced notice on the date of dam removal. Many small dams like the one in this study are removed with no prior notice and announcement. Many of the removal decisions, particularly for small dams in Pennsylvania, are made on an adhoc basis and are driven by the level of urgency (dam damage and safety hazard for the public) and availability of funds from state or local agencies for removal. Having prior knowledge of removal and funding for water quality monitoring will allow for a better assessment of pre- and post-dam removal effects.

Because of lack of resources, we were also unable to collect data for riparian soils and groundwaters for the first four months (August–November) post dam removal. This also happened to be an unusually active hydrologic period with large storms. Given the early nitrate-N increases we observed in stream water, it is very likely that similar rapid N changes also occurred in riparian groundwaters which we did not record. Our soil $\delta^{15}N$ data also revealed early and quick changes in the N status of soils. Given these early responses, it is important that water and soil monitoring be implemented early following dam removal, especially in hydrologically active conditions. High-frequency storm sampling or use of in-situ nitrate-N sensors (e.g., Aubert et al., 2016; Vaughan et al., 2017) may enable better characterization of storm-driven N flushing and episodic changes in N concentrations in stream and groundwaters.

In contrast to quick, short-term changes, this study also revealed that N changes (e.g., ammonium-N concentrations for W5) continue to occur more than 2 years after dam removal. Thus, there could be a staggered response across various riparian N reservoirs. This suggests that sampling will likely have to be continued for periods longer than 2 years, particularly for large dam removals to fully characterize the changes in riparian N conditions. Taller dam removals would result in larger water level drops and drainage and likely result in greater and/or different types of N cycle changes than those reported for this study. In addition, multiple dam removals, as opposed to single removals, could also have different impacts. Thus, dam removal effects on riparian N processes need to be evaluated for a range of dam removal conditions.

This study site was located in a predominantly agricultural watershed with elevated N concentrations and inputs from cropland fertilizer and animal waste. There also appeared to be watershed-wide declining trend in stream water N contrary to that hypothesized in this study. Thus, it is very likely that in developed watersheds such as the Chiques Creek, the effects of a single low-head dam removal on stream water N may not be very apparent and could be masked by elevated N agricultural and urban loadings, effects of N legacies (e.g., Chang et al., 2021) and broader regional and continental scale trends in N (e.g., Newcomer et al., 2021).

## 5. Conclusions and Broader Environmental Implications

To our knowledge, this is the first study that has explicitly investigated the effects of low-head dam removal on riparian N processing and leakage. We had hypothesized that dewatering of the riparian zones following milldam removal would result in loss of denitrification services, an increase in soil nitrification, and thus an increase in nitrate-N concentrations in soil, ground, and stream waters. Our observations yielded important results showing partial support for our hypotheses. Novel insights were derived with regard to how ammonium-N potentially influenced the status of N in riparian soils. Key conclusions were:

Dam removals and groundwater drainage resulted in partial loss of denitrification services in surficial riparian soils. This was supported by decreased denitrification rates and the soil $\delta^{15}N$ values. No significant changes in soil nitrification were observed during the two-year study period, but longer-term observation is recommended.

While nitrate-N concentrations were elevated in soil water, these concentrations were not observed in ground and stream waters, undermining our hypothesis of N leakage following dam removal and drainage. This suggests that surficial nitrate-N is either lost to denitrification at the deeper, but more dynamic groundwater interface, or is conserved through other N mechanisms such as DNRA. On the other hand, it is also possible that some N leakage could have occurred early in the study period during storms, which was not captured by our groundwater monitoring.

We observed accumulation of ammonium-N in near-stream riparian sediments upstream of the milldams. The anoxic, fine-grained, and C-rich sediments, a legacy of milldams, likely enhanced the potential for N being conserved as ammonium-N through the process of DNRA and/or suppression of nitrification.

Understanding how dam removals could impact riparian N processing is critical since riparian zones are an important management practice for nonpoint source pollution control. As a testament to the importance of riparian zones as a management tool for water quality, the Chesapeake Bay Program has set an annual goal to plant 900 miles of riparian buffers every year as well as established a final goal to develop 14,400 miles of riparian buffer by the year 2025 (Chesapeake Bay Program, 2016). The entire process requires substantial investments through federal, state and local funding sources estimated at hundreds of millions of dollars or greater (Alliance for the Bay, 2015). With such considerable investments of money and labor, it is important to assess the cumulative role of existing and removed dams on riparian buffer water quality effectiveness. While two-year results from this study suggest that effects of single, low-head dam removal on stream water nitrate-N exports are likely small, additional long-term (>5 years) mass-balance studies that include taller dam removals are needed for a comprehensive assessment. The potential for ammonium-N accumulation in riparian zones upstream of milldams and its source-sink behavior vis-à-vis stream waters also needs to be evaluated rigorously. Lastly, while this study did not evaluate plant-N uptake, dam removals and subsequent lowering of groundwater levels could also affect riparian plant N uptake (due to groundwater falling below the root zone) and needs to be assessed thoroughly. In closing, milldams and their anthropogenic legacies have significant effects on riparian ecosystems and the choices we make (e.g., dam removals) could have important consequences for water quality.

## Data Availability Statement

All data used in this manuscript is posted on Hydroshare.org (DOI: https://www.hydroshare.org/resource/fe747d3511d84df7bb2856e2c7e37c27/).

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